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Title: Uranium and Plutonium Isotopic Ratio Methods using FRAM

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Uranium and Plutonium Isotopic Ratio Methods using FRAM

Fixed energy

Response function

Analysis with

Multiple efficiencies

Pete Karpius

FRAM Isotopic Analysis Software

- FRAM is a powerful isotopic analysis code that has the same underlying theory as MGA.
- Fixed-energy Response-function Analysis with Multiple efficiencies
- Self-calibration using several gamma-ray peaks
- Analyze gamma ray data from 30keV to >1MeV of HPGe planar, HPGe Coaxial, or CdTe detector
- The key difference between the MGA family of codes and FRAM is the use of **user-editable analysis parameters** in the latter.

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What Can FRAM Do?

- Control data acquisition
- Automatically analyze newly acquired data
- **Analyze previously acquired data**
- Provide information on the quality of the analysis
- Facilitate analysis in unusual situations
  - Non-standard energy calibrations
  - Gamma rays from non-SNM isotopes
  - Poor spectra (within limits)

Controlling Data Acquisition

- FRAM is not MCA software; it requires additional software to control basic detector settings
  - High Voltage
  - Pulse Shaping
- FRAM can be used to set acquisition times in real or live time
- FRAM shows you the spectrum as it is acquired if FRAM is controlling acquisition
  - Count time
  - Number of counts in the (moveable) cursor channel

Isotopic Ratio Principle

$$\frac{N^{i}}{N^{k}} = \frac{C(E^{i}_{j})}{C(E^{k}_{l})} \times \frac{T^{i}_{1/2}}{T^{k}_{1/2}} \times \frac{BR^{k}_{l}}{BR^{i}_{j}} \times \frac{RE(E_{l})}{RE(E_{j})}$$

N^i = Number of atoms of isotope i

$C(E^i_j)$ = Photopeak area of gamma ray j with energy E_j emitted from isotope i

$T^i_{1/2}$ = Half-life of isotope i

BR^i_j = Branching ratio (gamma rays/disintegration) of gamma ray j from isotope i

$RE(E_j)$ = Relative detection efficiency of gamma ray with energy E_j. Includes detector efficiency, measurement geometry, sample self-absorption, and attenuation in materials between the sample and detector.

Relative Efficiency

- General Expression for Peak Area Count Rate

$$\mathbf{C} = \frac{\mathbf{N} \times \mathbf{ln\ 2} \times \mathbf{BR} \times \mathbf{RE}}{\mathbf{T}_{\mathbf{1/2}}}$$

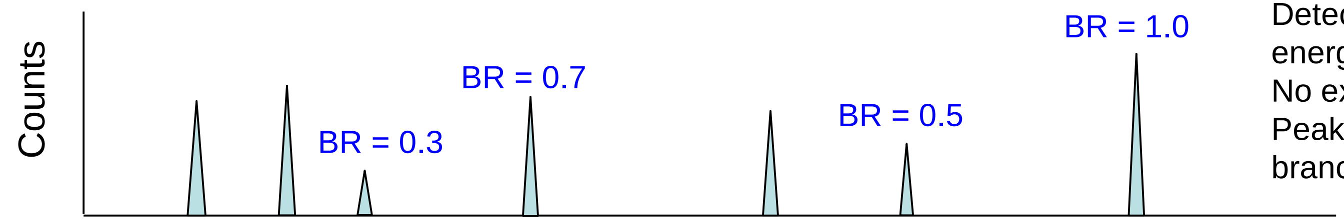
- Rearrange

$$\frac{\mathbf{C}}{\mathbf{BR}} = \left\{ \frac{\mathbf{N} \times \mathbf{ln\ 2}}{\mathbf{T}_{\mathbf{1/2}}} \right\} \times \mathbf{RE}$$

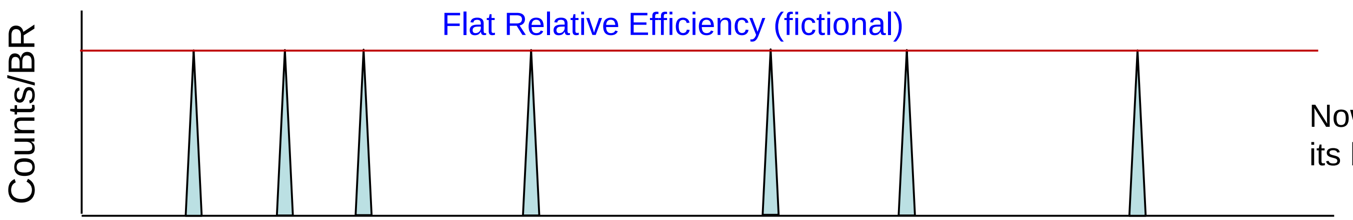
- Relative efficiency is proportional to count rate and branching ratio

$$\mathrm{RE} \propto \frac{\mathrm{C}}{\mathrm{BR}}$$

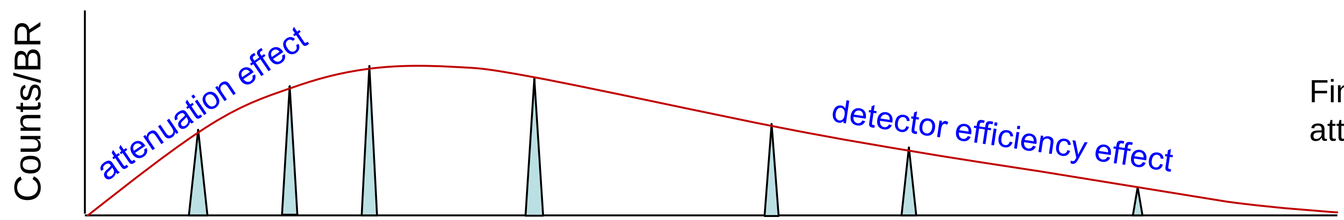
Relative Efficiency (Fictional Spectrum)



Detector has same efficiency at all energies (fictional).
No external or self attenuation.
Peak height only depends on branching ratios.

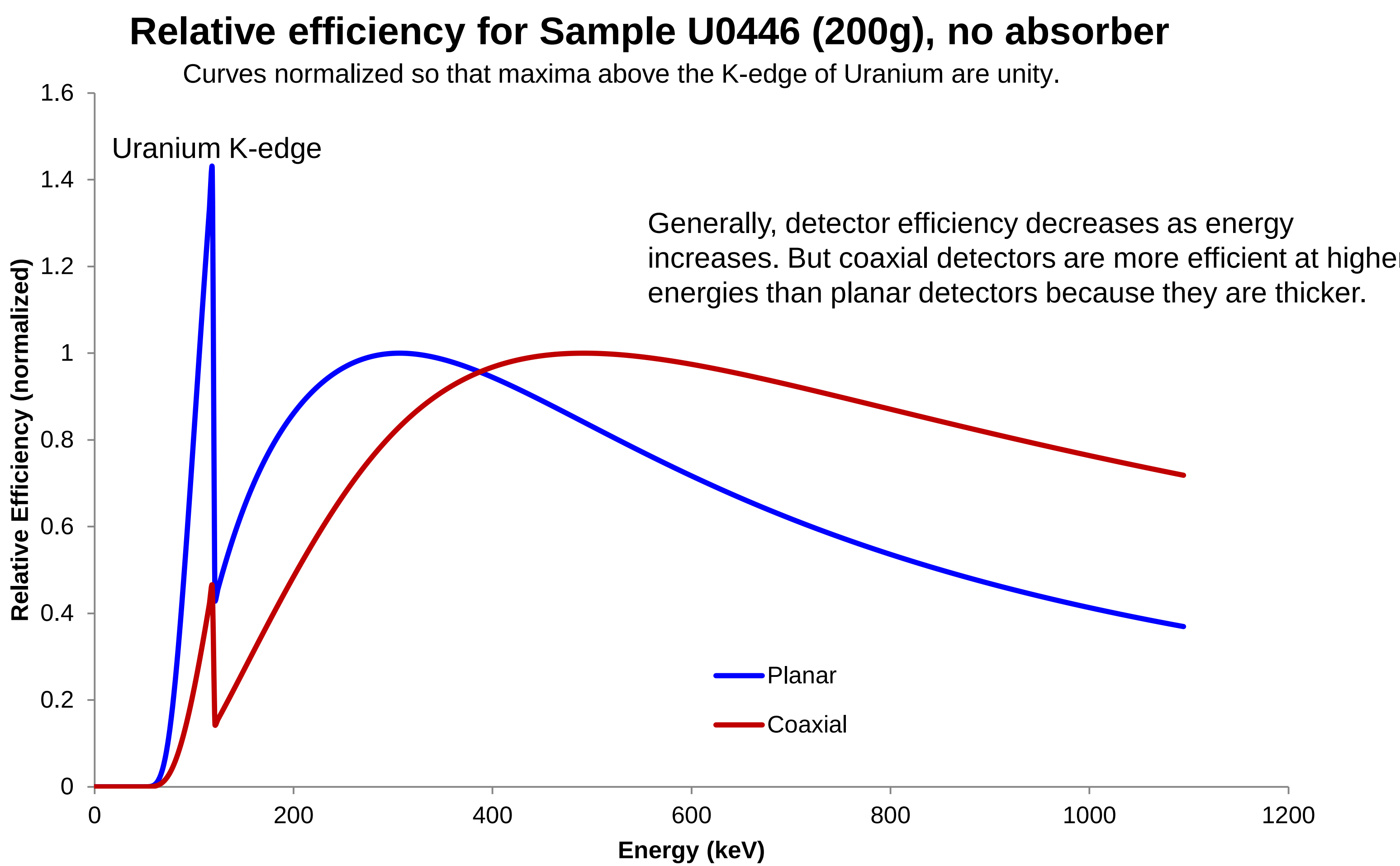


Now <u>normalize</u> each peak area by its branching ratio.

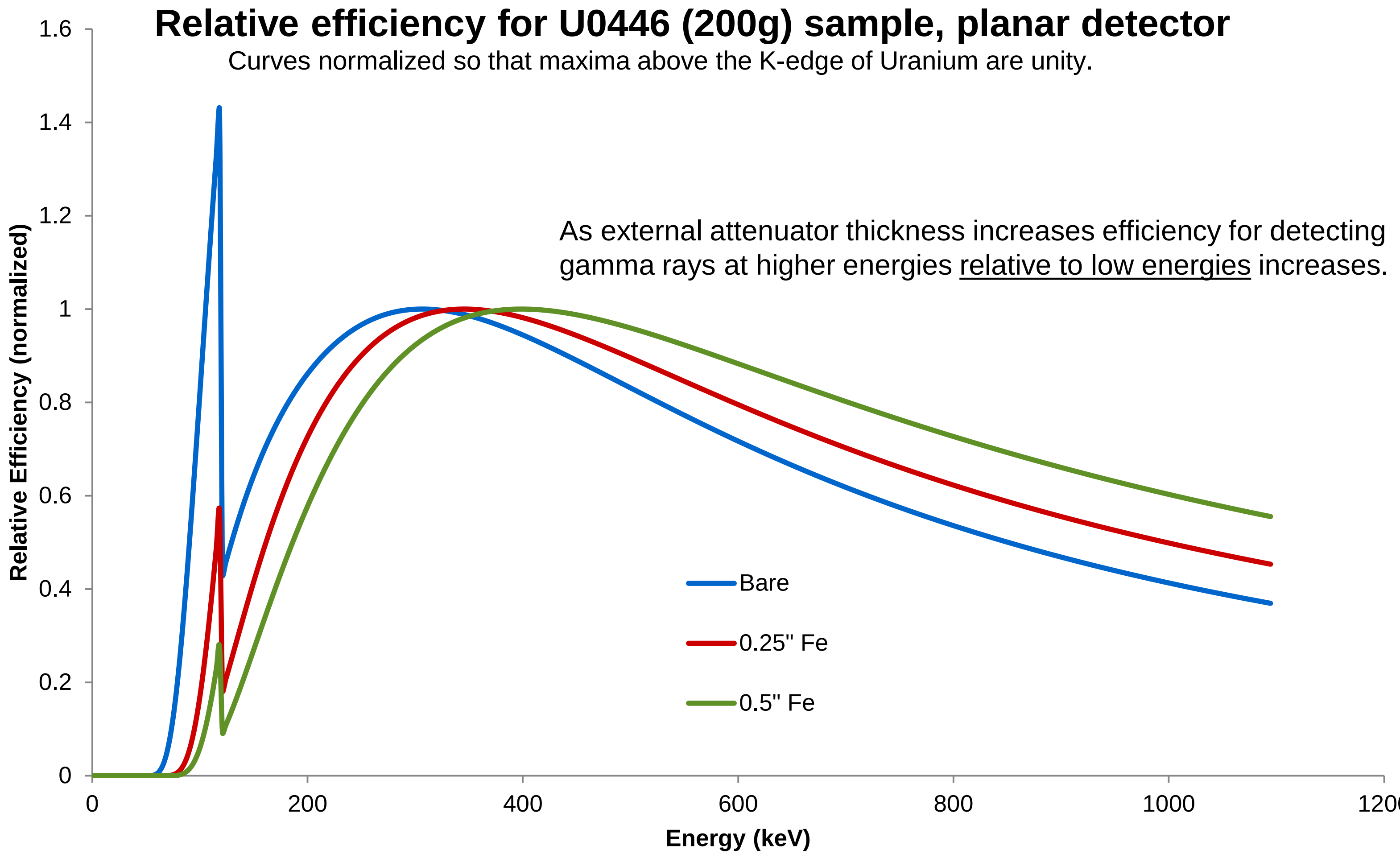


Finally, consider the real effects of attenuation and detector efficiency.

Relative Efficiency Depends on Detector Efficiency



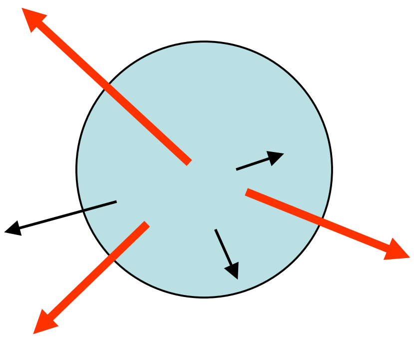
Relative Efficiency Depends on External Attenuation



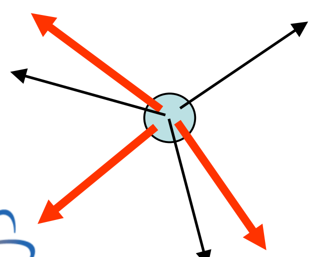
Relative Efficiency Depends on Self Attenuation

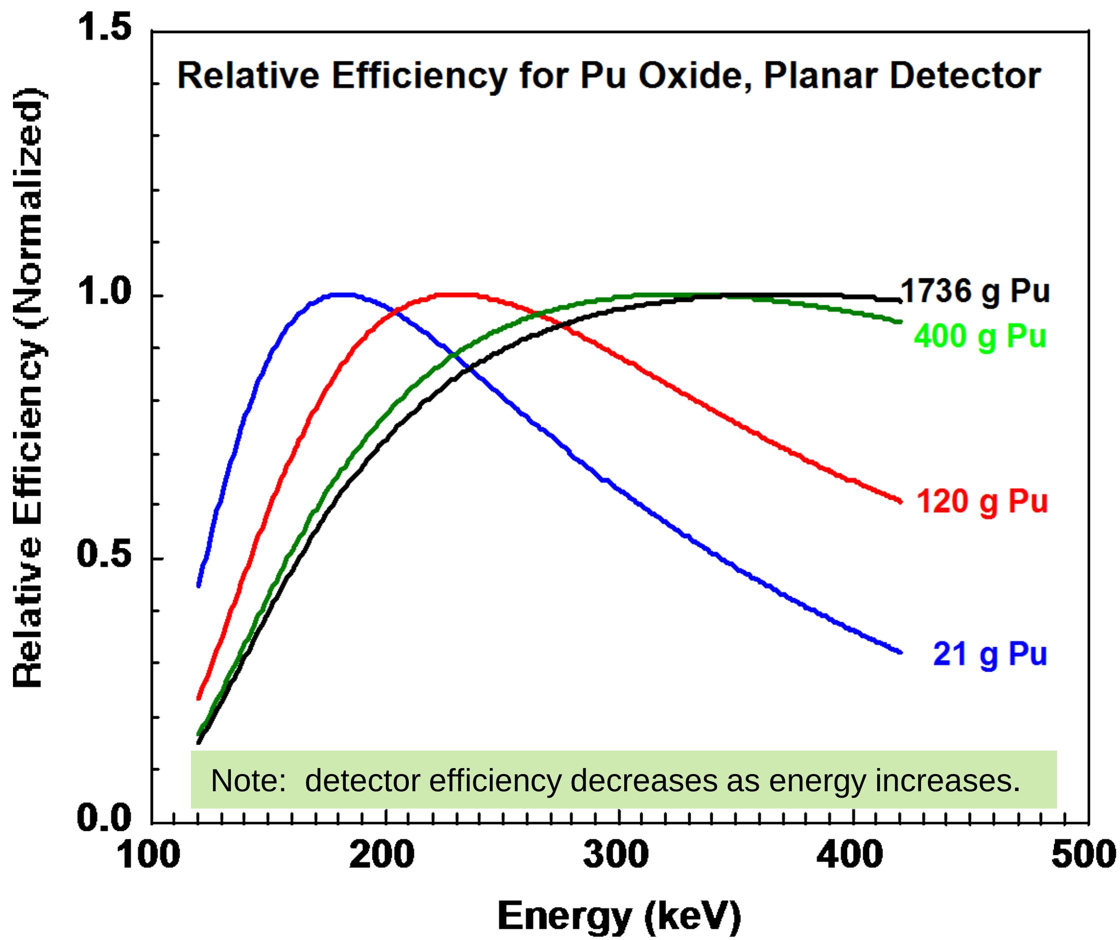
= Low E γ Rays
= High E γ Rays

Large Mass Item



Small Mass Item





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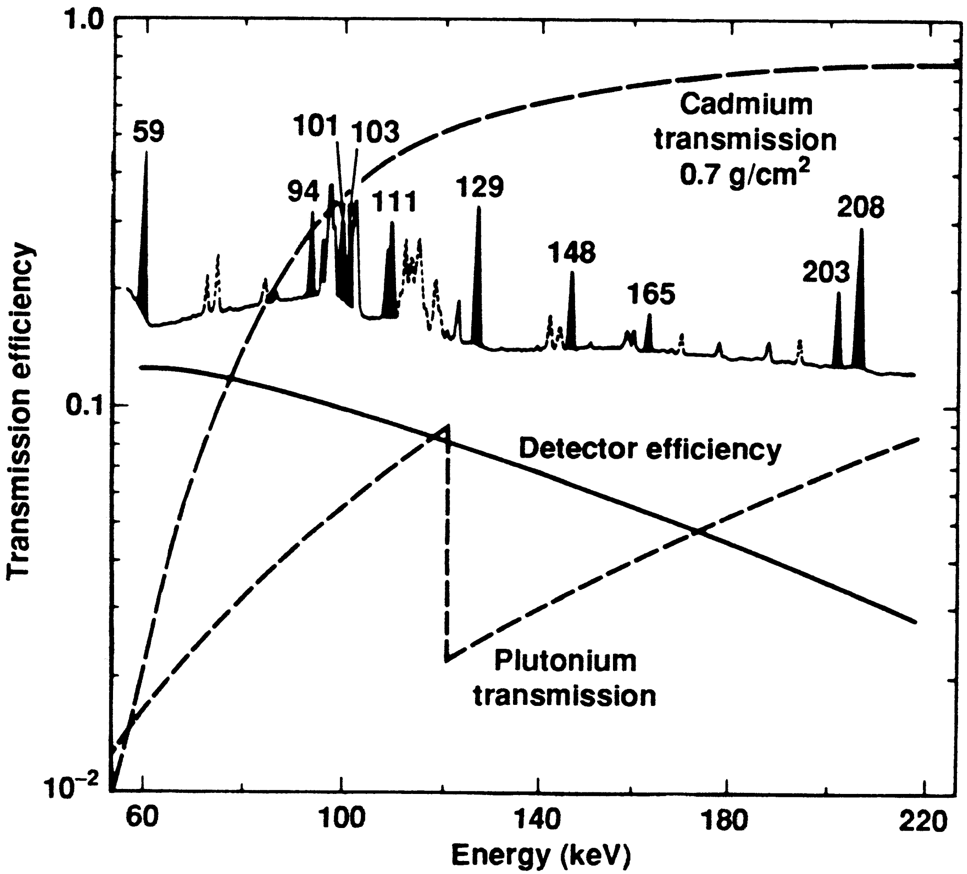
Relative Efficiency Curve Components



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Independence of Analysis

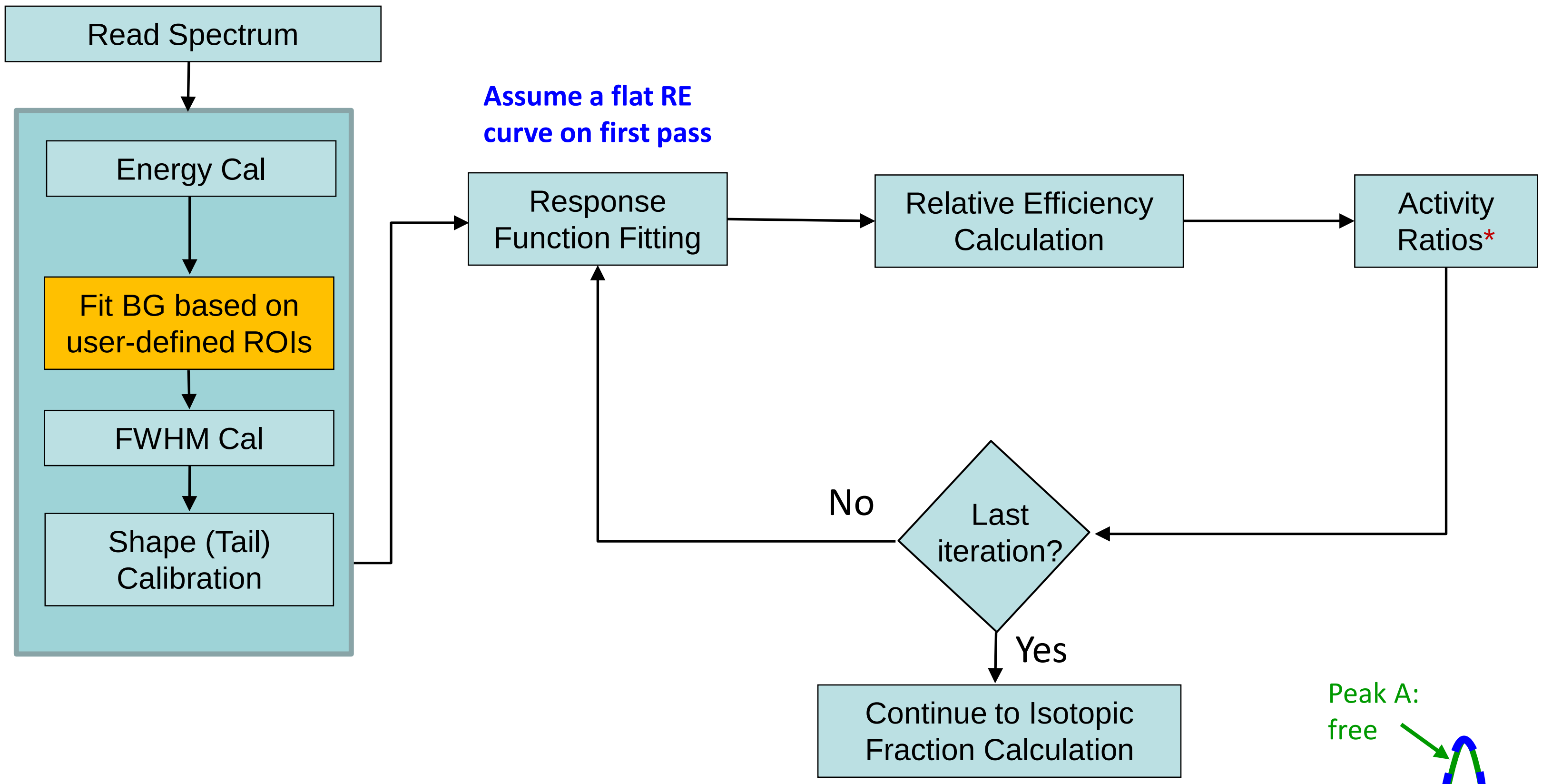
- Using **relative** efficiency and isotope **ratios** makes analysis independent of:
  - **Item Characteristics**
    - Size
    - Shape
    - Physical and Chemical Composition
    - Packaging
    - Filtering
  - **Data Acquisition Limitations**
    - Pulse Pile-up
    - Deadtime

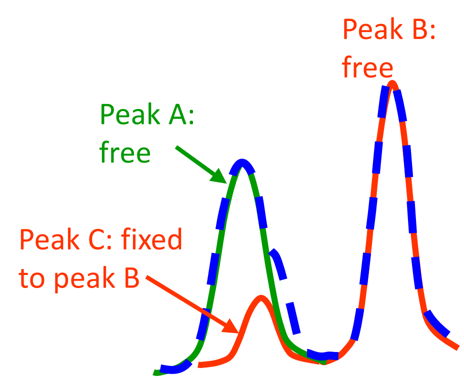
NO CALIBRATION IS NEEDED!

FRAM Methodology

- **Internal Calibration**
  - Energy
  - FWHM
  - Peak Shape (Tailing Parameters)
- **Analysis**
  - Background continuum
  - Peak areas (Response Functions)
  - Relative Efficiency
  - Isotopic Ratio Calculation
  - Isotopic Mass Fraction Calculation

FRAM Top-Level Flow to Calculation of Activity Ratios

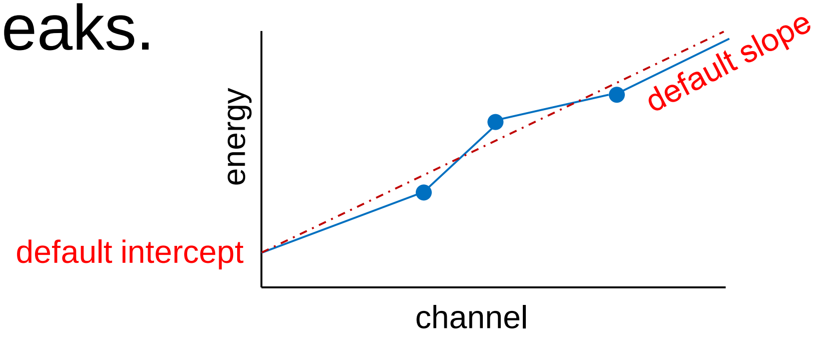




*Note: activity ratio calculations are needed in the iterations on the RE curve as peak stripping of often applied using multiple nuclides, which make up the data for the RE curve.

Internal Calibration

- **Energy** -- Piecewise linear calibration between designated peaks.



- **FWHM**^{2} = $A_1 + A_2{*}E + A_3{*}E^{-1}$ fit to FWHM of designated peaks

- **Shape** -- Gaussian with exponential tail -- tail parameters found from residuals on designated peaks after subtracting Gaussian component.

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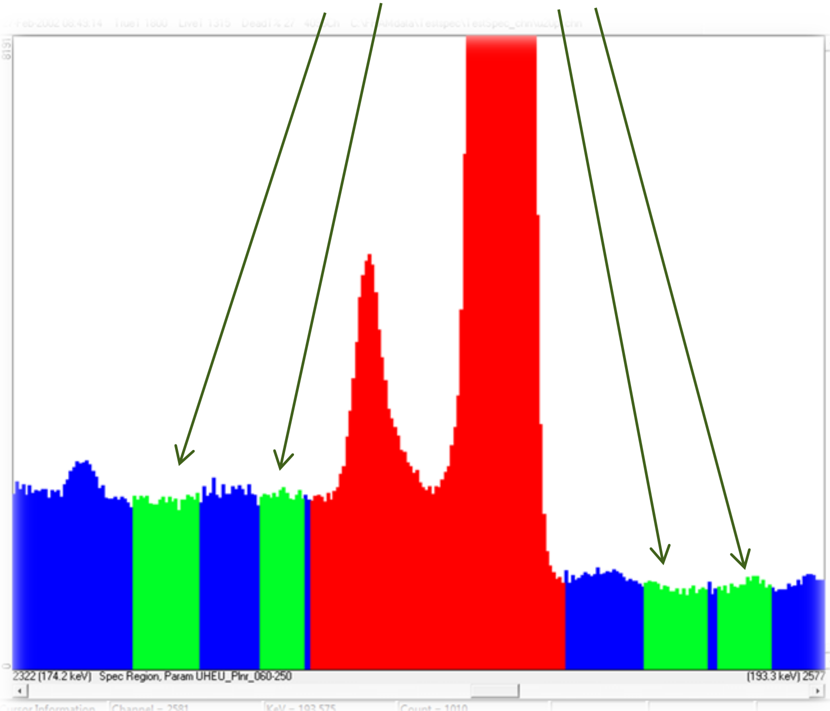
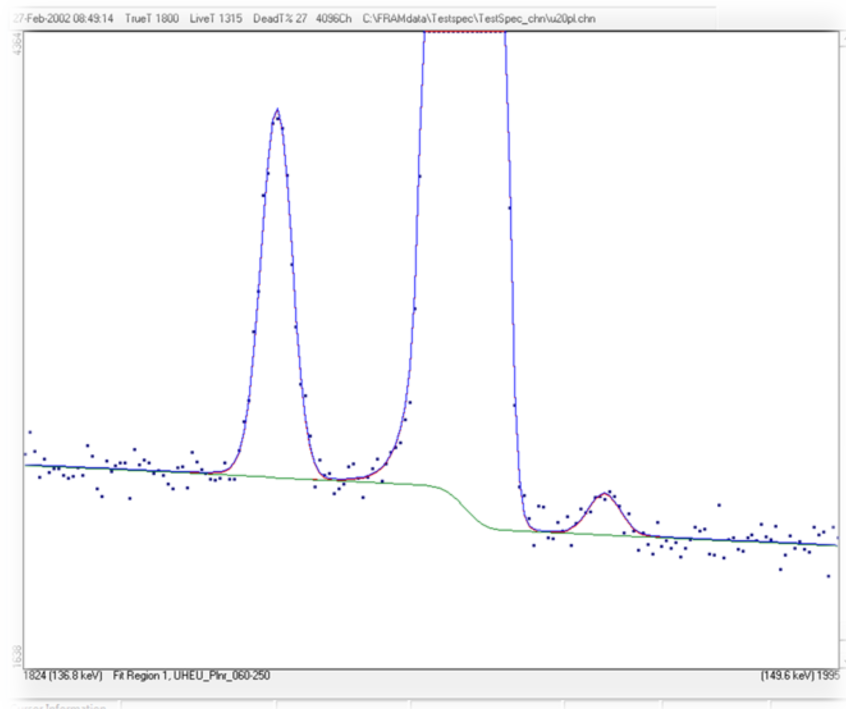
Background Continuum

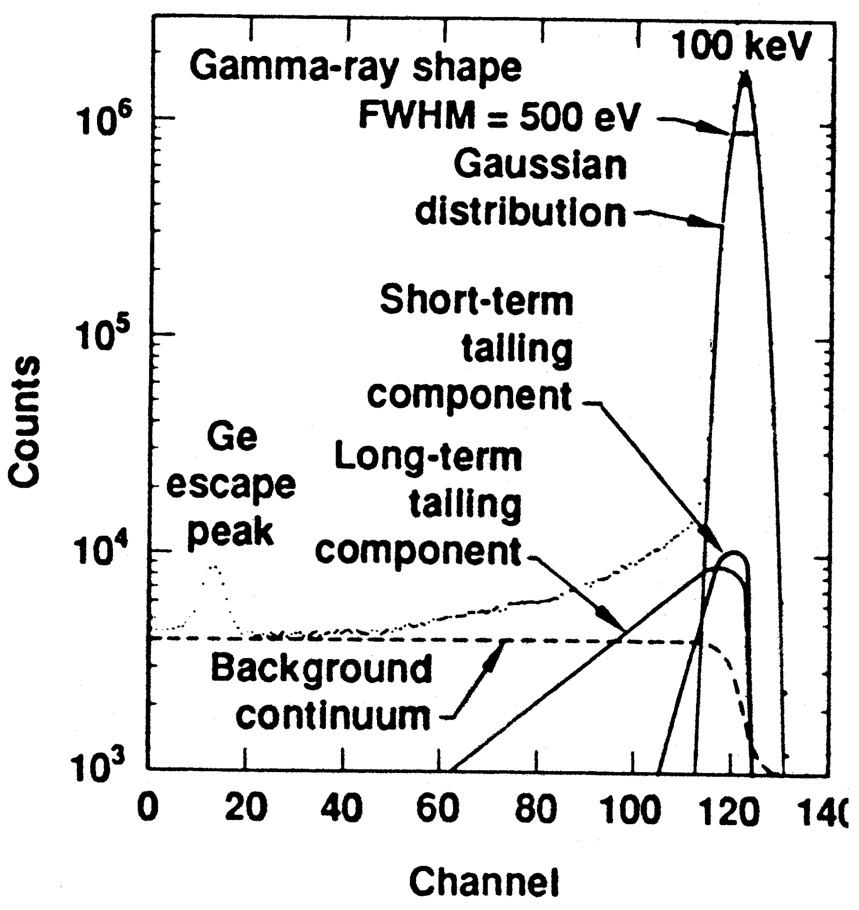
Continuum Regions of Interest



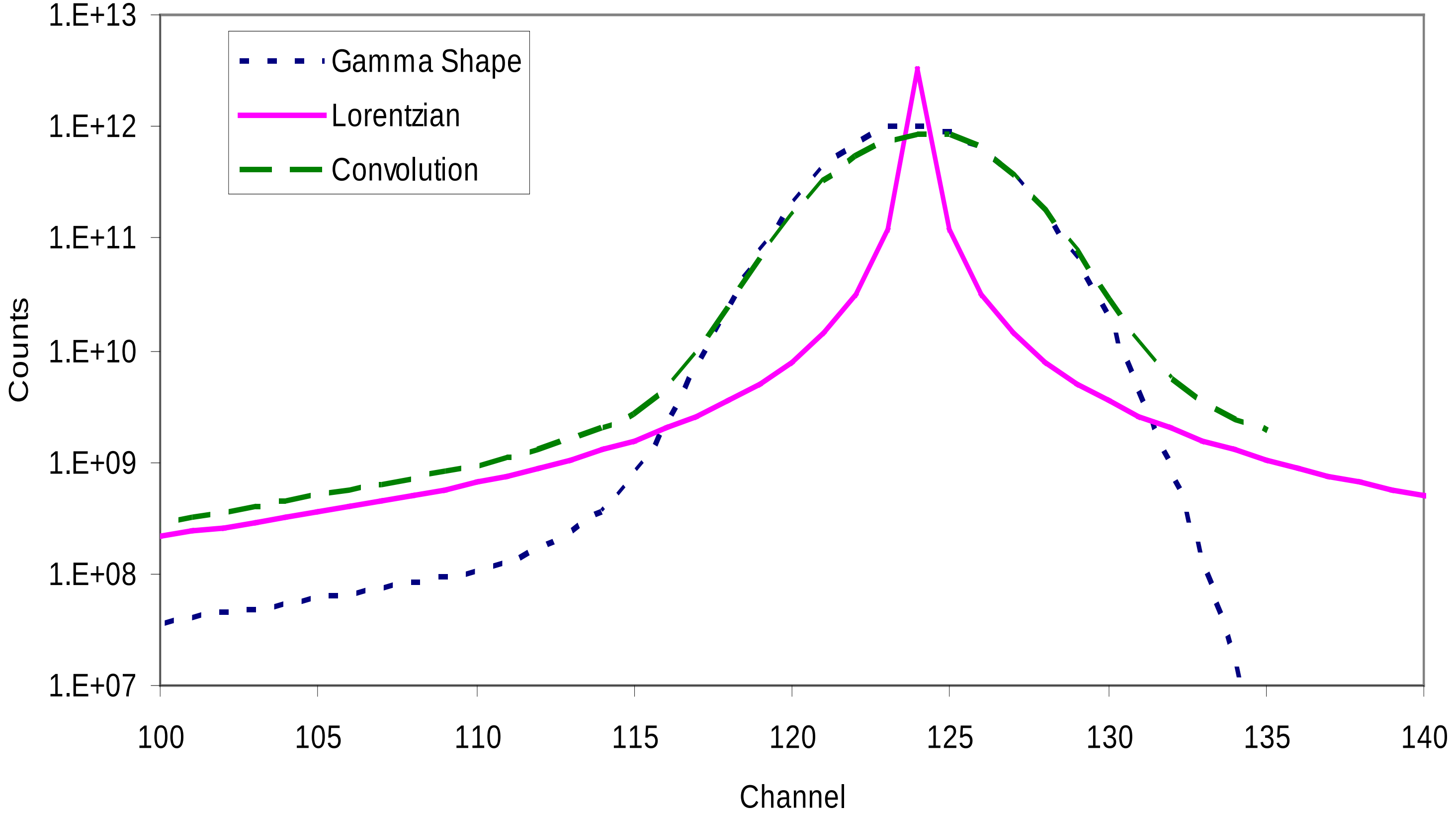
Calculated Step Function



Gamma-Ray Peak Fitting



X-Ray Peak Shape Convolution (Voigt)



Relative Efficiency Models

- Empirical or Physical
- The empirical model works well with good statistics within a range of continuity.
- The physical model is based on our understanding of radiation shielding physics
- It requires more knowledge
  - Density of the nuclear material
  - Major shielding materials and thicknesses
- The physical model can cross efficiency discontinuities (such as the K edge of Pu)

Empirical Relative Efficiency Curve

$$Y = C_1 + C_2E^{-2} + C_3(lnE) + C_4(lnE)^2 + C_5(lnE)^3 + C_i + C_jE^{-1}$$

Y	=	log of ratio of (peak area/branching ratio)
E	=	energy
C_i	=	normalization for isotopes beyond the first one
C_j	=	normalization for different relative efficiency curves

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Physical Efficiency Model

$$RE_i = \frac{1 - e^{-\mu_{Pu} \cdot x_{Pu}}}{\mu_{Pu} \cdot x_{Pu}} \cdot \left(\prod_k e^{-\mu_k \cdot x_k} \right) \cdot I_i \cdot e^{\beta_i / E} \cdot DetEff \cdot E^b c^{1/E}$$

Self Attenuation

Hörl Function
(adjustment for variation in detector efficiency and matrix)

Intensity of i^{th} Isotope

External Attenuation
(where k = Pb, Cd, Fe)

Heterogeneity Factor
(associates additional efficiency functions beyond the first one)

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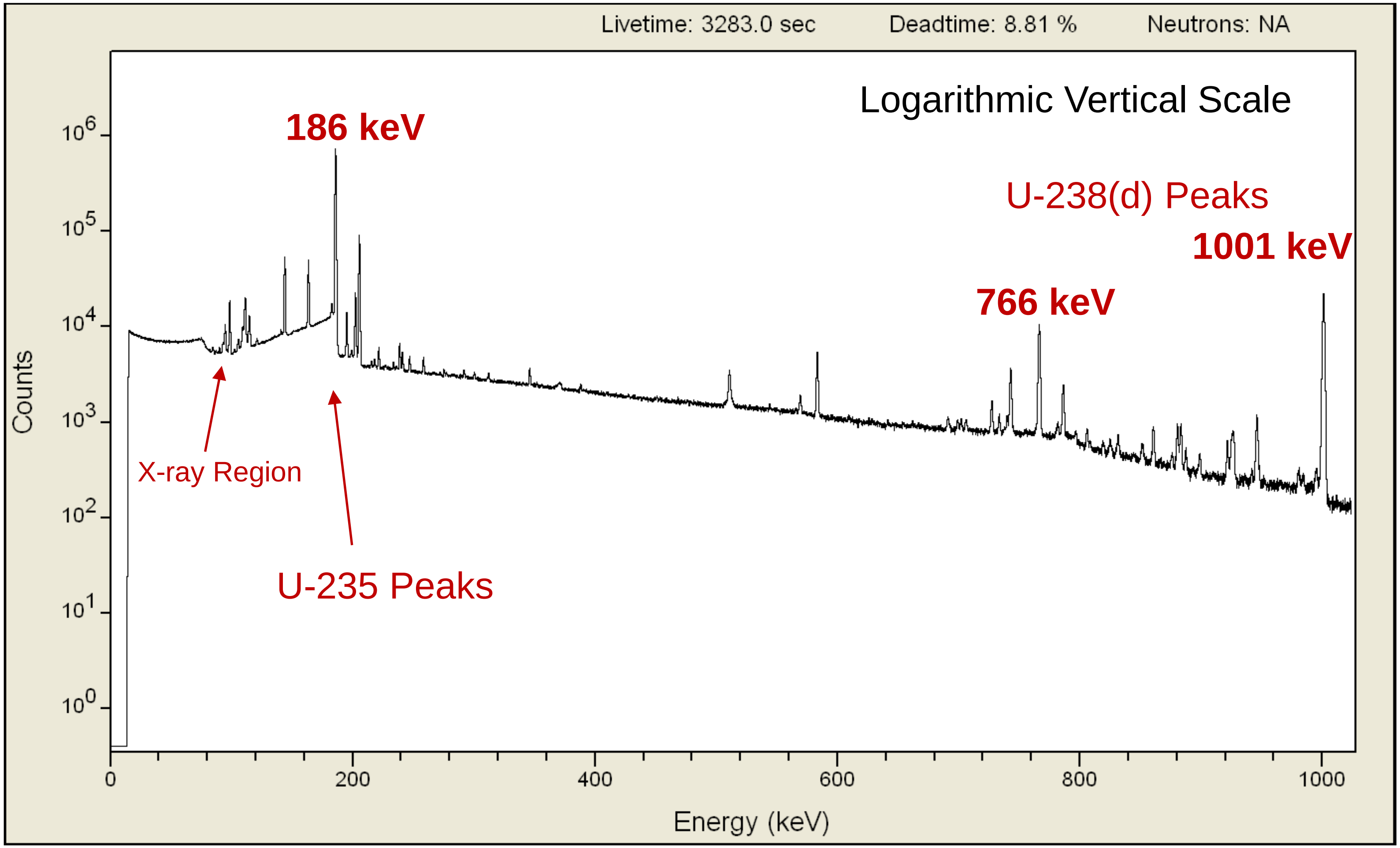
Uranium Gamma-Ray Spectrum (1-MeV Range)



Uranium Isotopic Composition from Peak Ratios

- From the spectrum, measure the ratios:
  - $^{238}f/^{235}f \leftrightarrow$ Isotopic Ratio($^{238}U/^{235}U$)
  - $^{234}f/^{235}f \leftrightarrow$ Isotopic Ratio($^{234}U/^{235}U$)
- Then calculate the ^{235}U fraction using the constraint:

$$1 = {}^{238}f + {}^{235}f + {}^{234}f + {}^{236}f$$

i.e.

$$\boxed{{}^{235}f = \left(1 + {}^{238}f/\,{}^{235}f + {}^{234}f/\,{}^{235}f + ({}^{236}f/\,{}^{235}f\,)\right)^{-1}}$$

- Use the ^{235}U fraction to solve for ^{238}f and ^{234}f .

^{236}U Estimate by Correlation

- Determine relative content of ^{236}U from a <u>correlation function</u> based on the fractions of the other uranium isotopes.
- Example form of a correlation function to determine the fraction of ^{236}U from the fractions of ^{235}U and ^{238}U:

$$^{236}f = A \cdot ({}^{235}f^{B} \cdot {}^{238}f^{C})$$

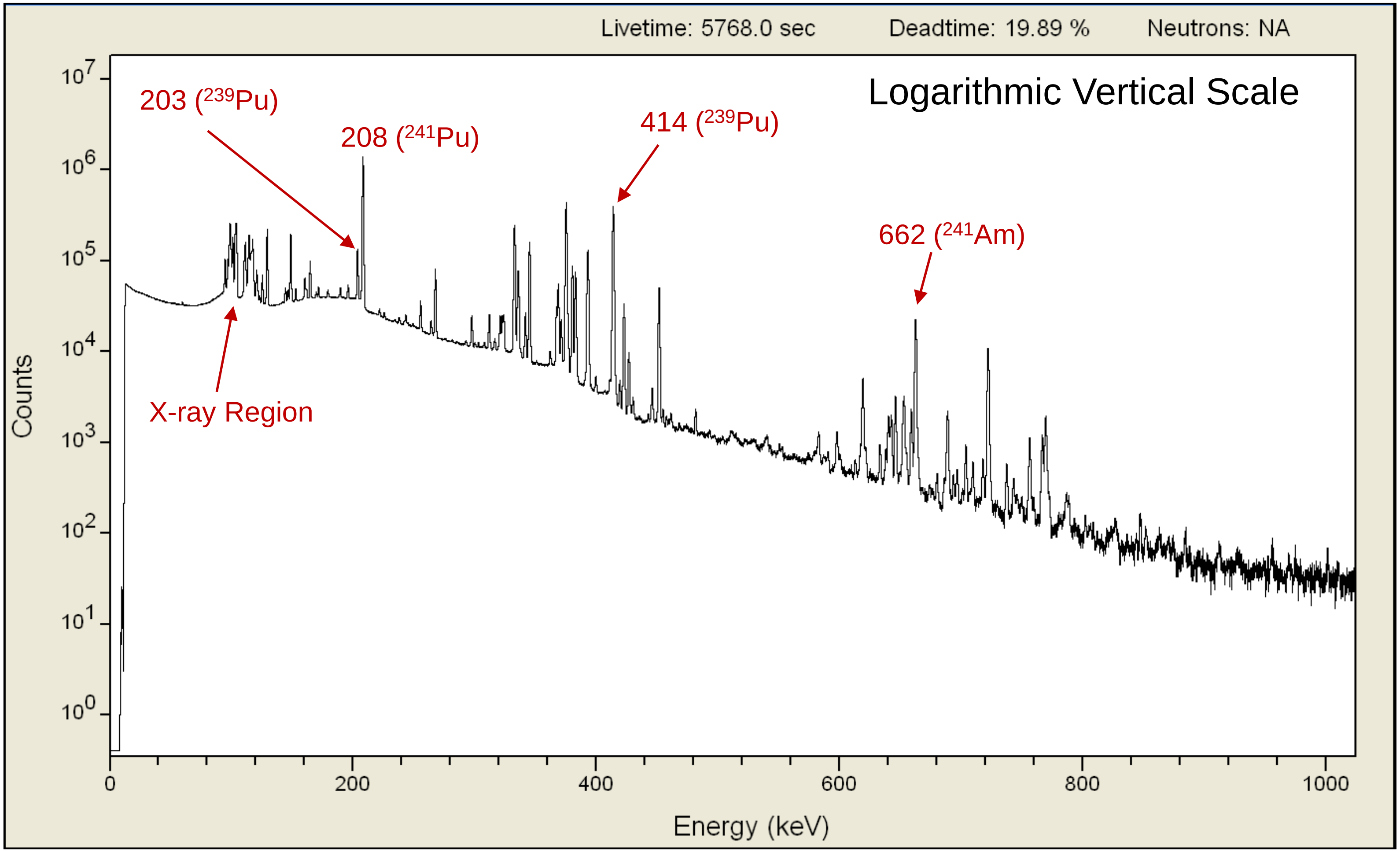
Ref. Los Alamos National Laboratory Document LA-UR-11-03005

- Once we have the fraction of ^{236}U, we can put it back into:

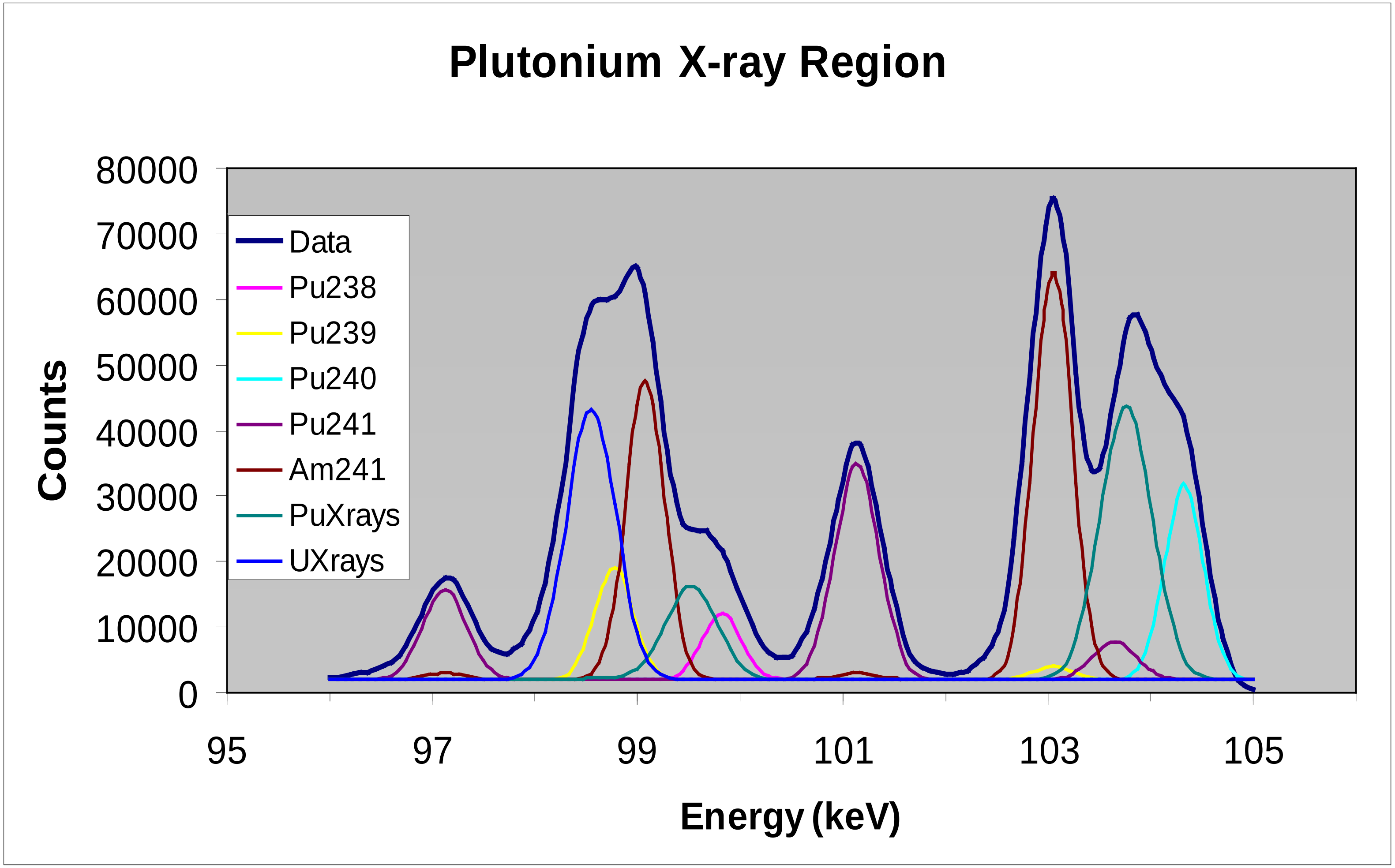
$$^{238}f + {}^{235}f + {}^{234}f + {}^{236}f \geq 1$$

…and then renormalize ^{238}f, ^{235}f, and ^{234}f by this new sum.

Plutonium Gamma-Ray Spectrum (1-MeV Range)



Plutonium Response Function Analysis



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Plutonium Isotopic Fractions from Isotopic Ratios

- Measure isotopic ratios : ${}^{238}f/{}^{239}f$

  ${}^{240}f/{}^{239}f$

  ${}^{241}f/{}^{239}f$

  $({}^{242}f/{}^{239}f)$

- Sum of all fractions must equal 1:

$$1 = {}^{238}f + {}^{239}f + {}^{240}f + {}^{241}f + ({}^{242}f)$$

- Divide by ${}^{239}f$ and then solve for ${}^{239}f$:

$$\boxed{{}^{239}f = [{}^{238}f/{}^{239}f + 1 + {}^{240}f/{}^{239}f + {}^{241}f/{}^{239}f + ({}^{242}f/{}^{239}f)]^{-1}}$$

^{242}Pu Estimate by Correlation

- Determine relative content of ^{242}Pu from a correlation function based on the fractions of the other plutonium isotopes:

$$^{242}\mathbf{Pu} = 53[^{240}\mathbf{Pu}]\mathbf{x}[^{241}\mathbf{Pu}]/[^{239}\mathbf{Pu}]^2$$

Gunnink, Nucl. Matl. Mgmt. 9, (2), 83-93 (1980)

Typical form of correlation functions used by MGA (*& FRAM*):

$$^{242}\mathbf{Pu} = A\times[\ (^{238}\mathbf{Pu})^B \times (^{239}\mathbf{Pu})^C \times (^{240}\mathbf{Pu})^D \times (^{241}\mathbf{Pu} + {}^{241}\mathit{Am})^E]$$

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Note: We renormalize the isotopic fractions as we did with ^{236}U.

Requirements for Isotopic Analysis

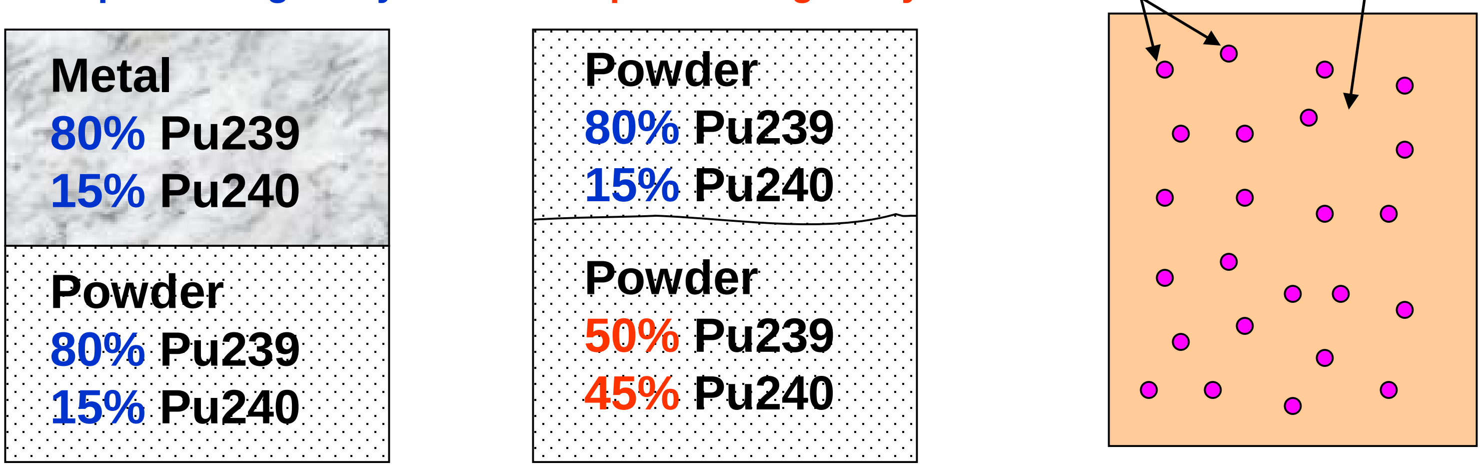
- **Isotopic Homogeneity**
  - All plutonium (or uranium) must have the same isotopic composition even if it is not physically or chemically alike
- **Pu and Am must have same spatial distribution**

Isotopic Homogeneity

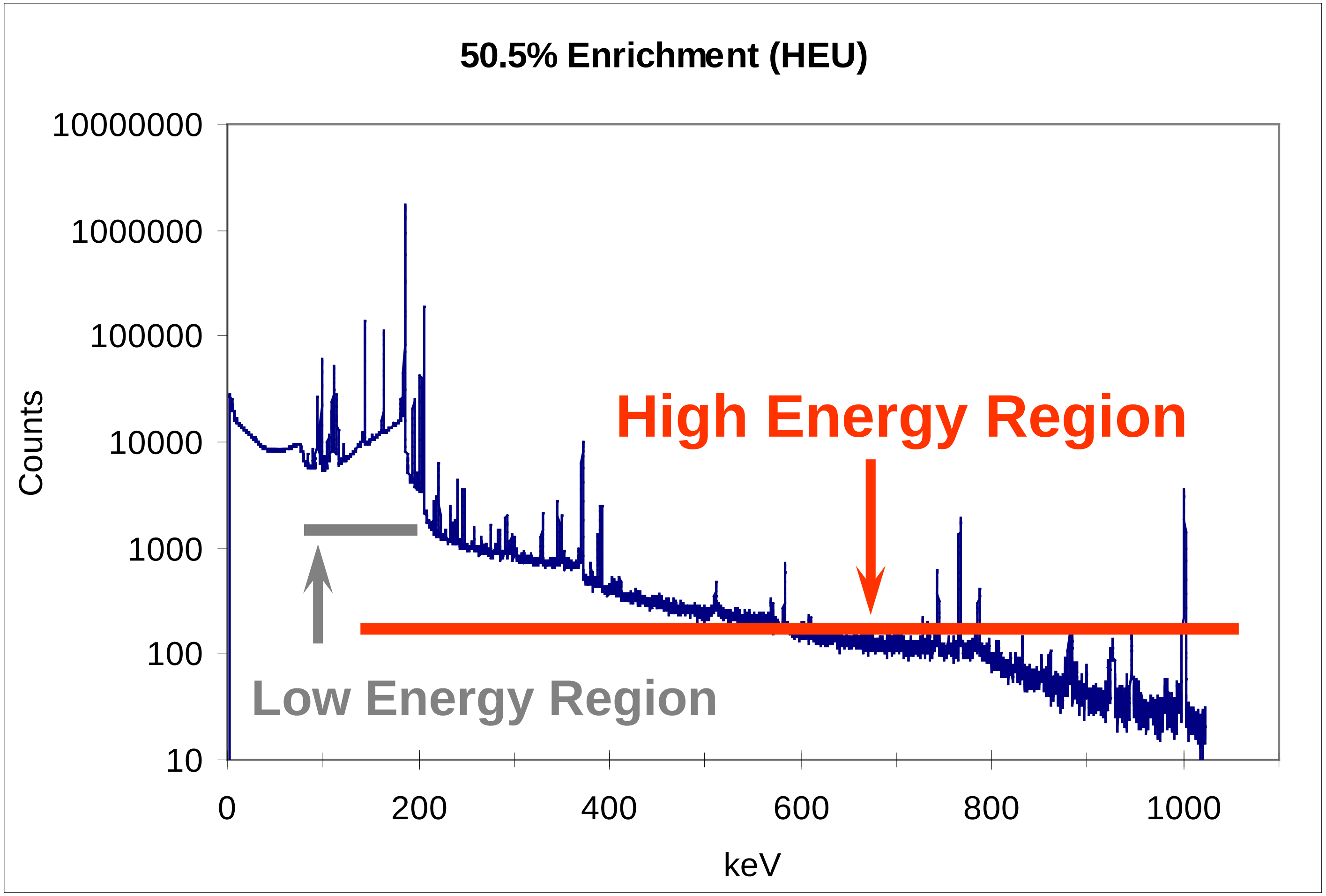
- The isotopic composition must be uniform throughout the item, even if it is chemically or physically non-uniform.
  - One exception is that FRAM can analyze physically uniform, but isotopically heterogeneous, matrices of plutonium and ^{241}Am.

Isotopic Homogeneity: **Isotopic Heterogeneity:** **The Exception:** **Pu-metal in Am241 salt**

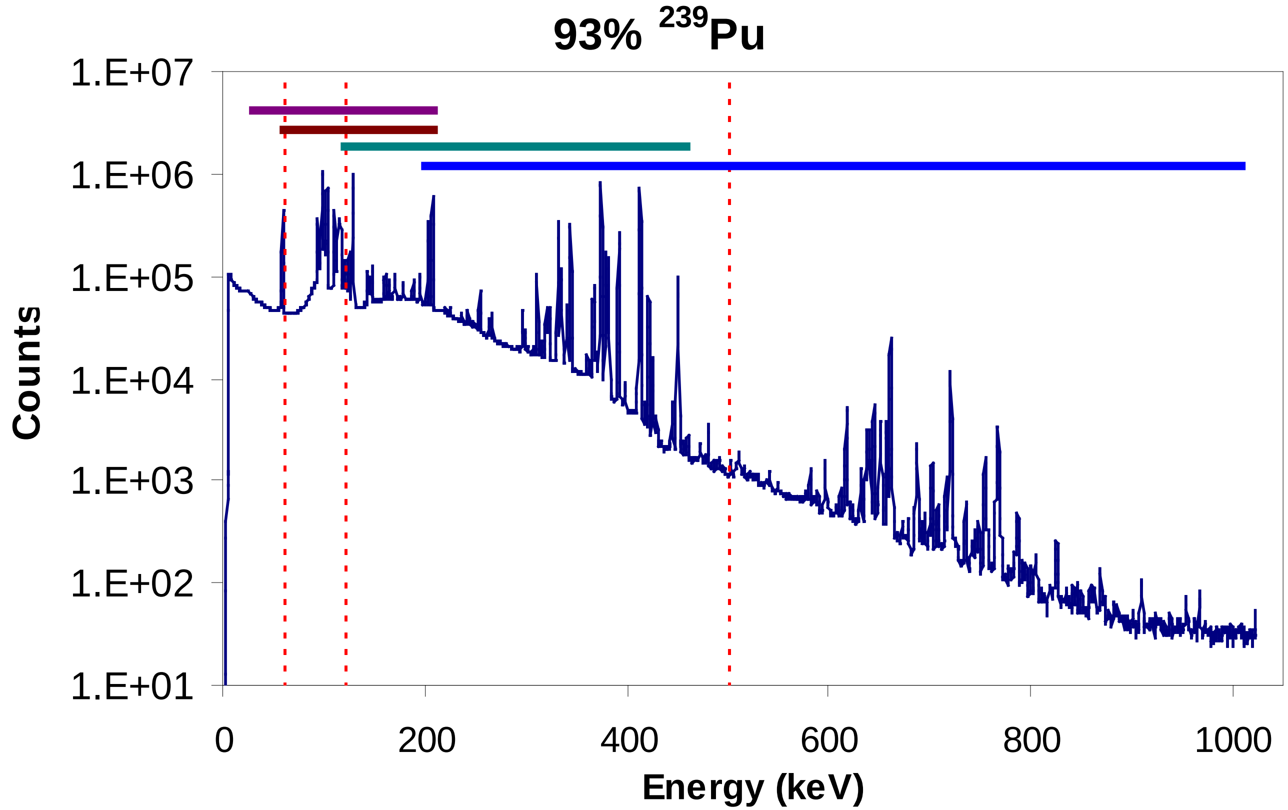
example:



Uranium Analysis Regions of FRAM



Plutonium Analysis Regions of FRAM



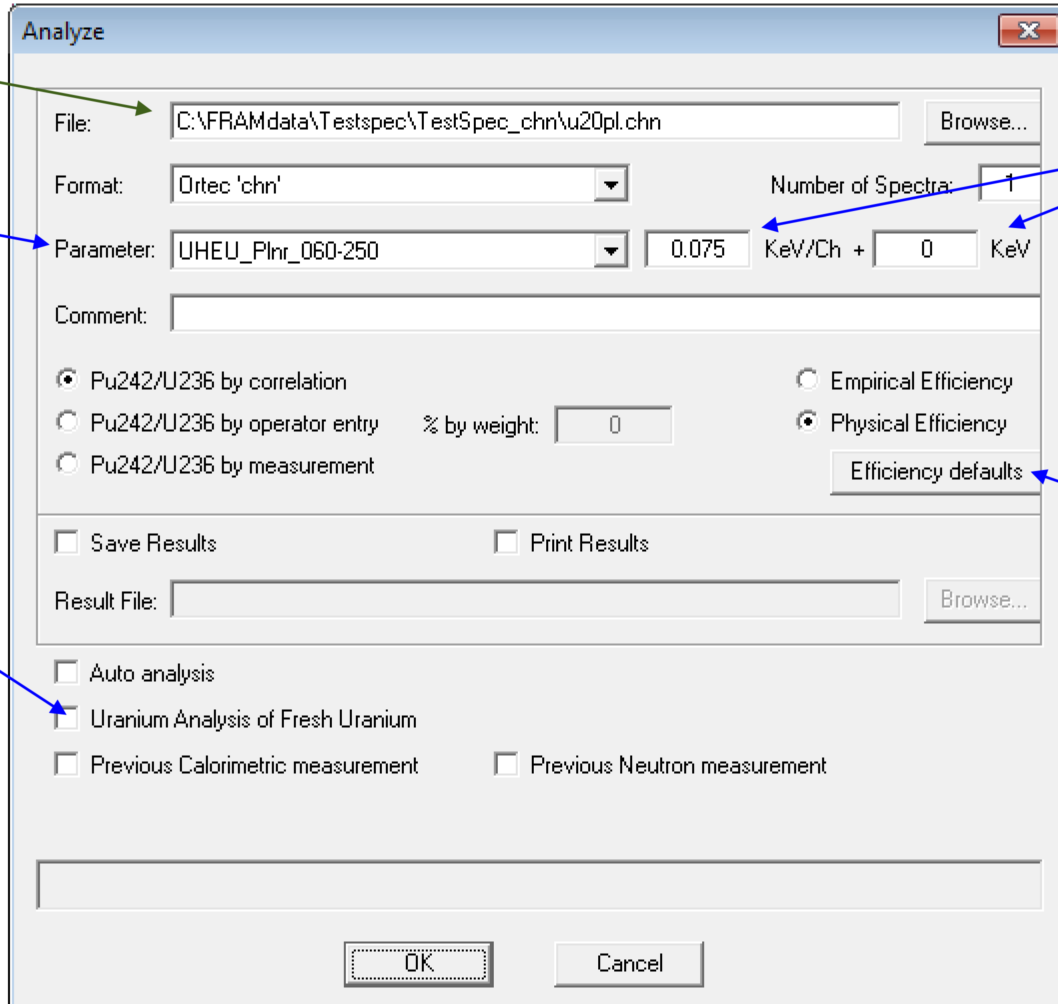
The four overlapping analytical regions that FRAM normally uses for the analysis are shown as four thick horizontal bars above the spectrum.

FRAM Analyze Window

Spectrum File

Parameter set

For uranium not in secular equilibrium



Default gain and offset

Efficiency Curve Option

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FRAM (Short) Results Window

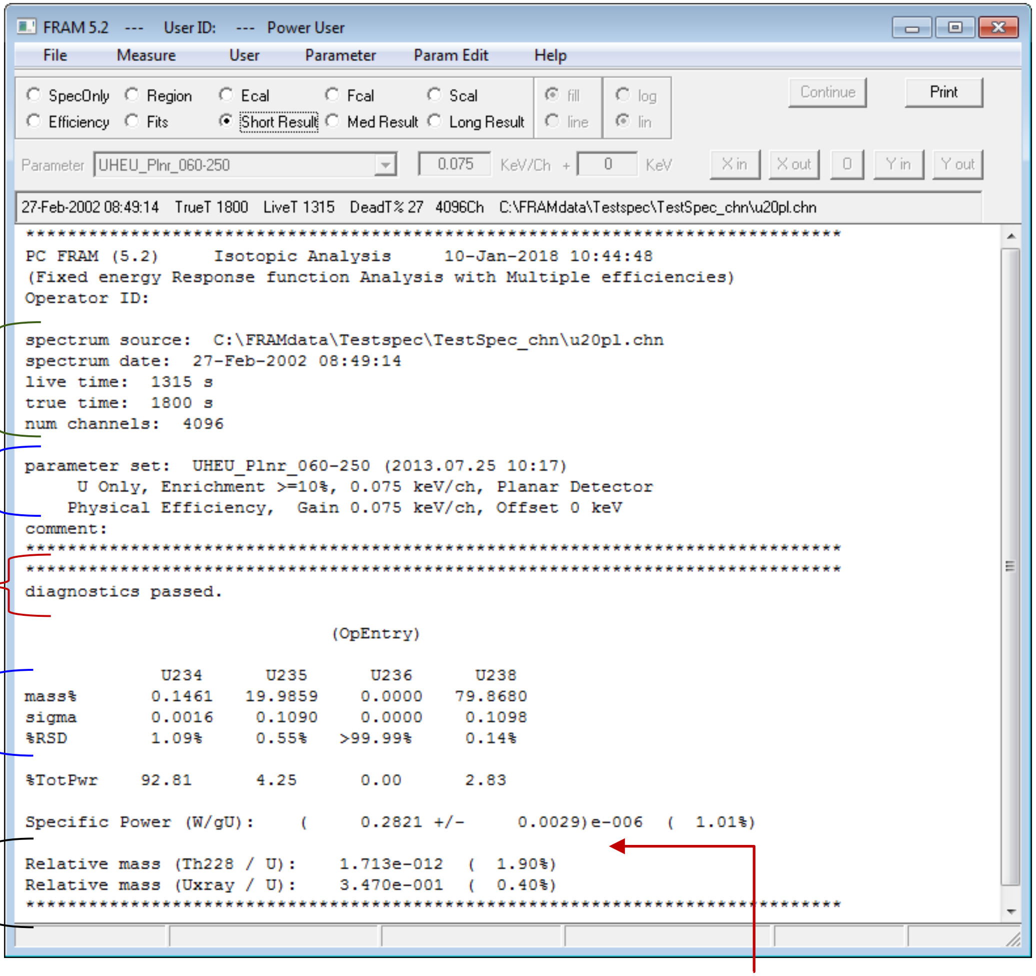
Raw data info

Parameter set

Errors/Warnings

Isotopic Fractions

Relative masses of other nuclides to U or Pu



Note: For Plutonium you will see "time since ^{241}Am separation" in this location

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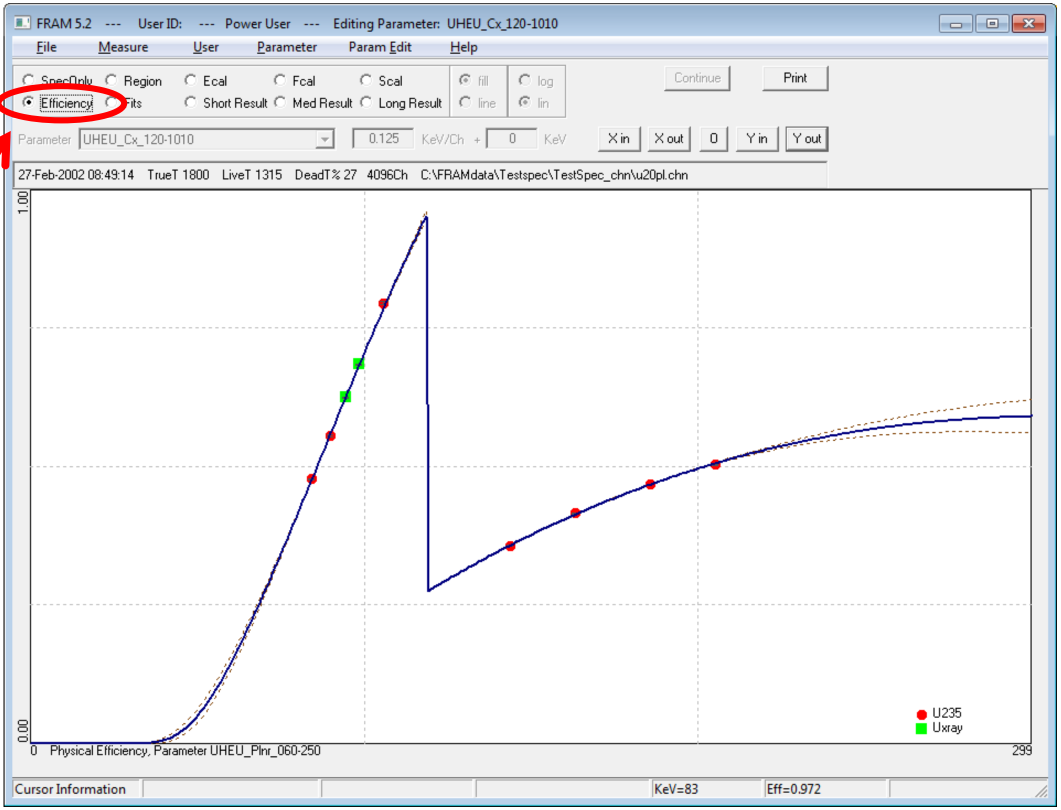
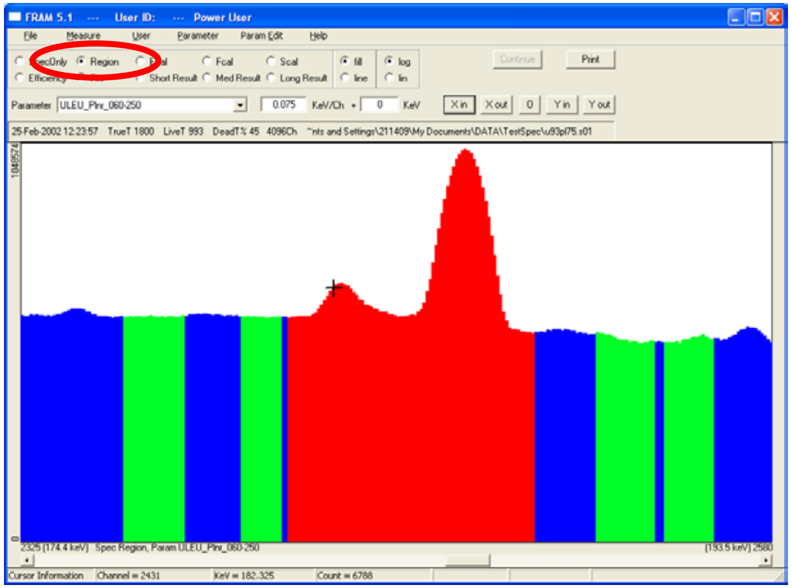
FRAM Diagnostics

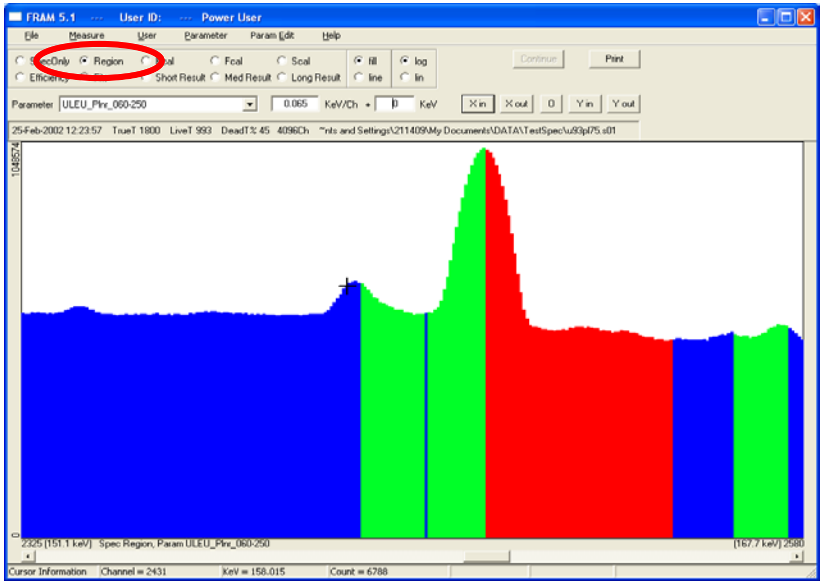
- FRAM provides diagnostic tools for the user to check the quality of the analysis
- **Diagnostic Tools**
  - Spectrum
  - Fits
  - Relative Efficiency
  - Results



Region: Checking the GAIN

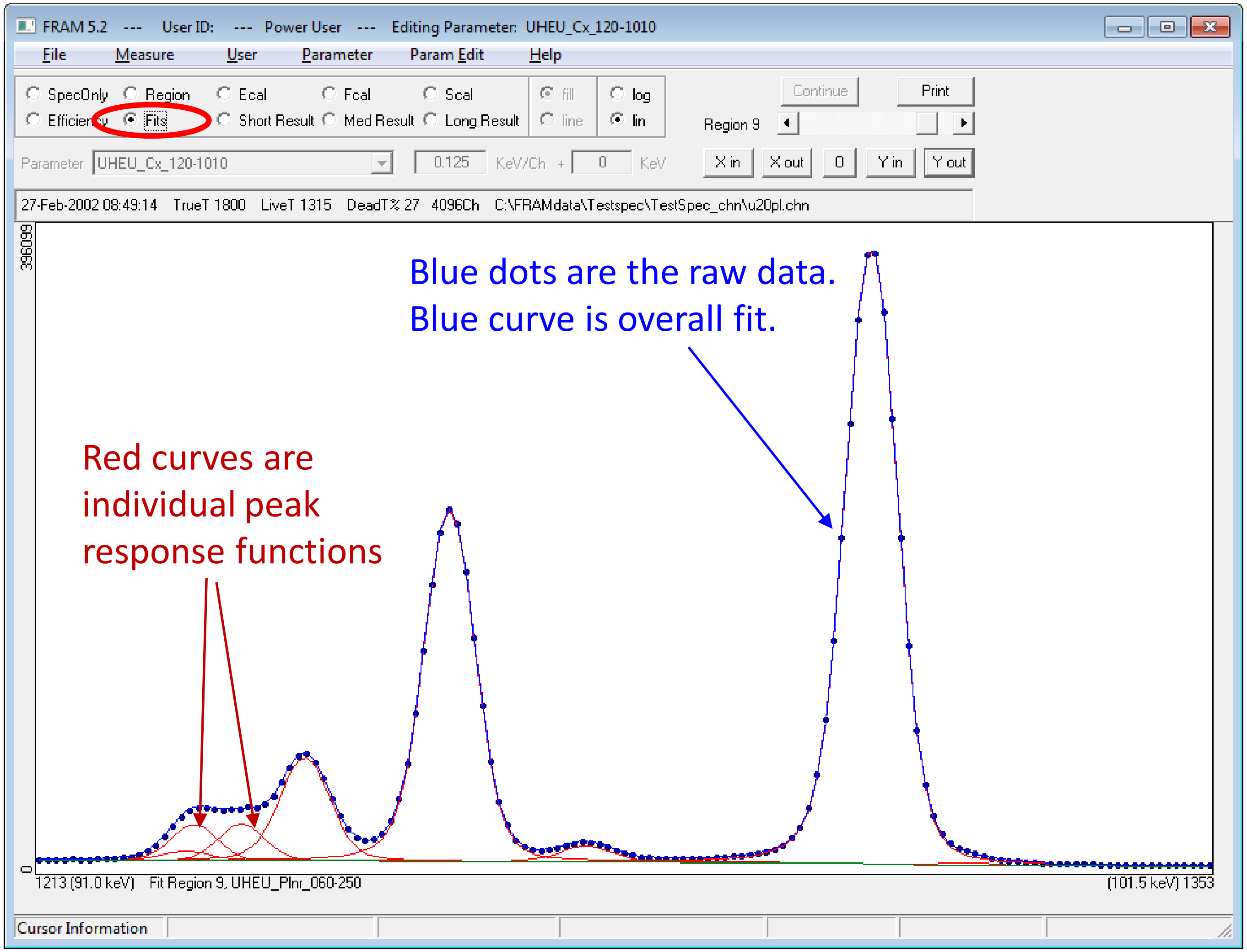


ROIs aligned with peaks
Default ECAL is OK



ROIs NOT aligned with peaks
Check default ECAL

Fits: Sum of All Peaks in Region



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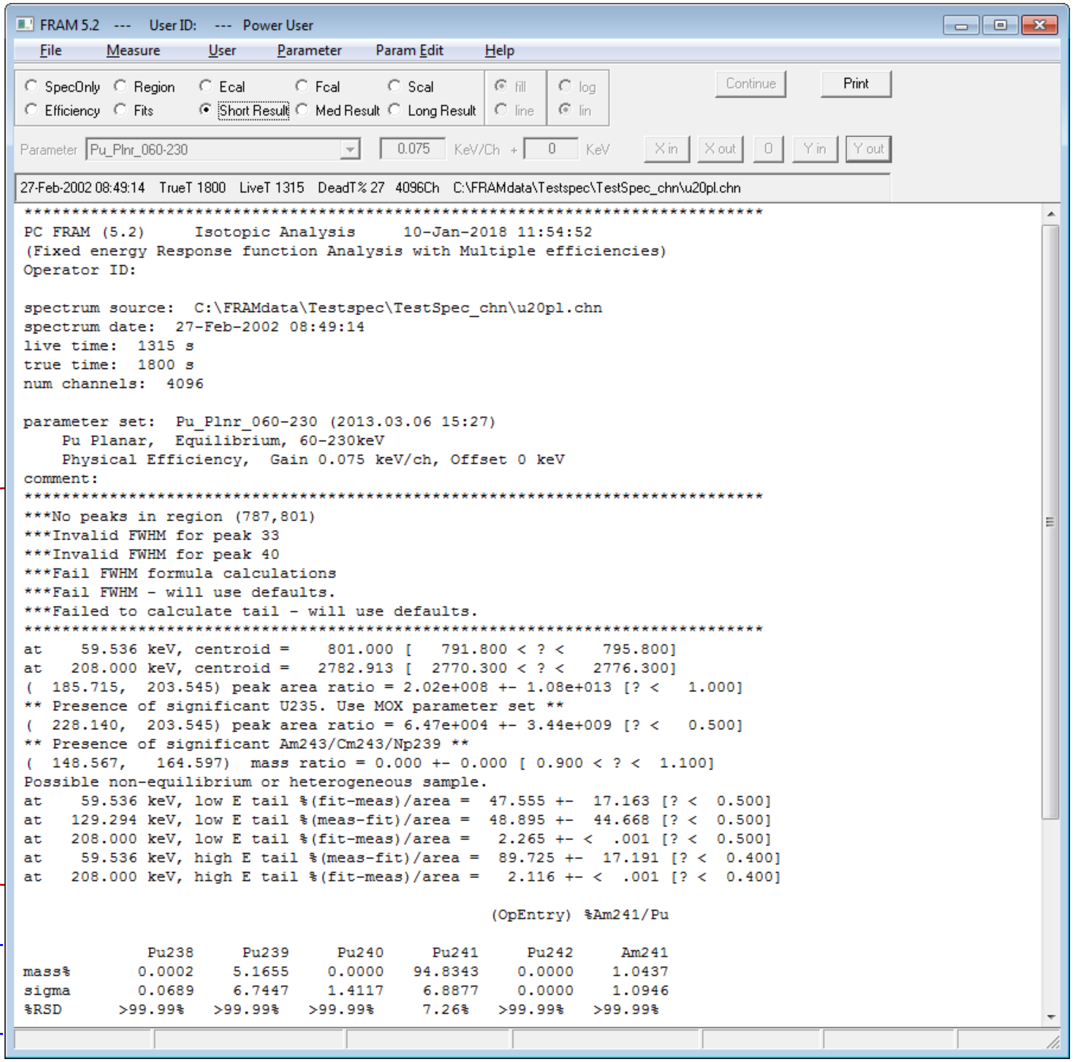
FRAM Results (Example: Major Errors)

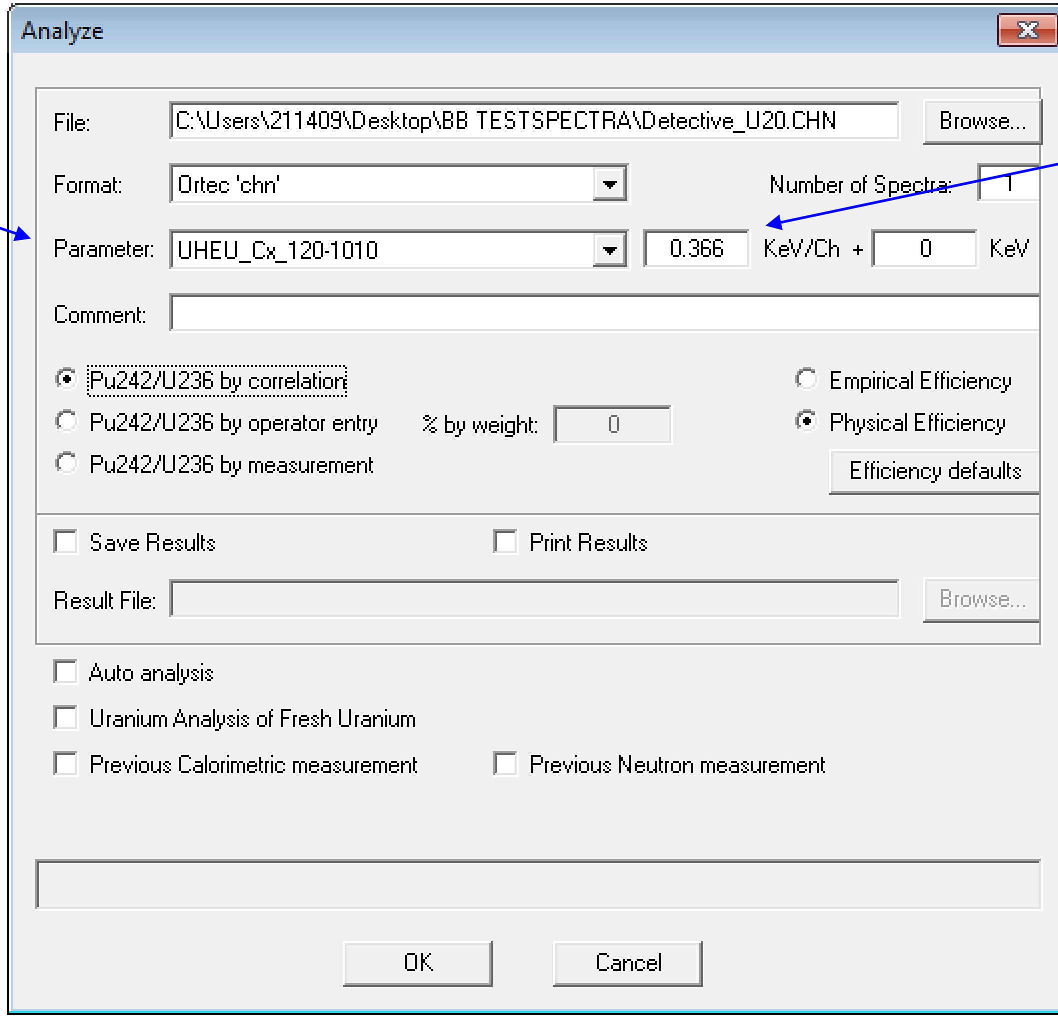
Now we have tons of error messages and warnings

Isotopic Fractions are nonsensical



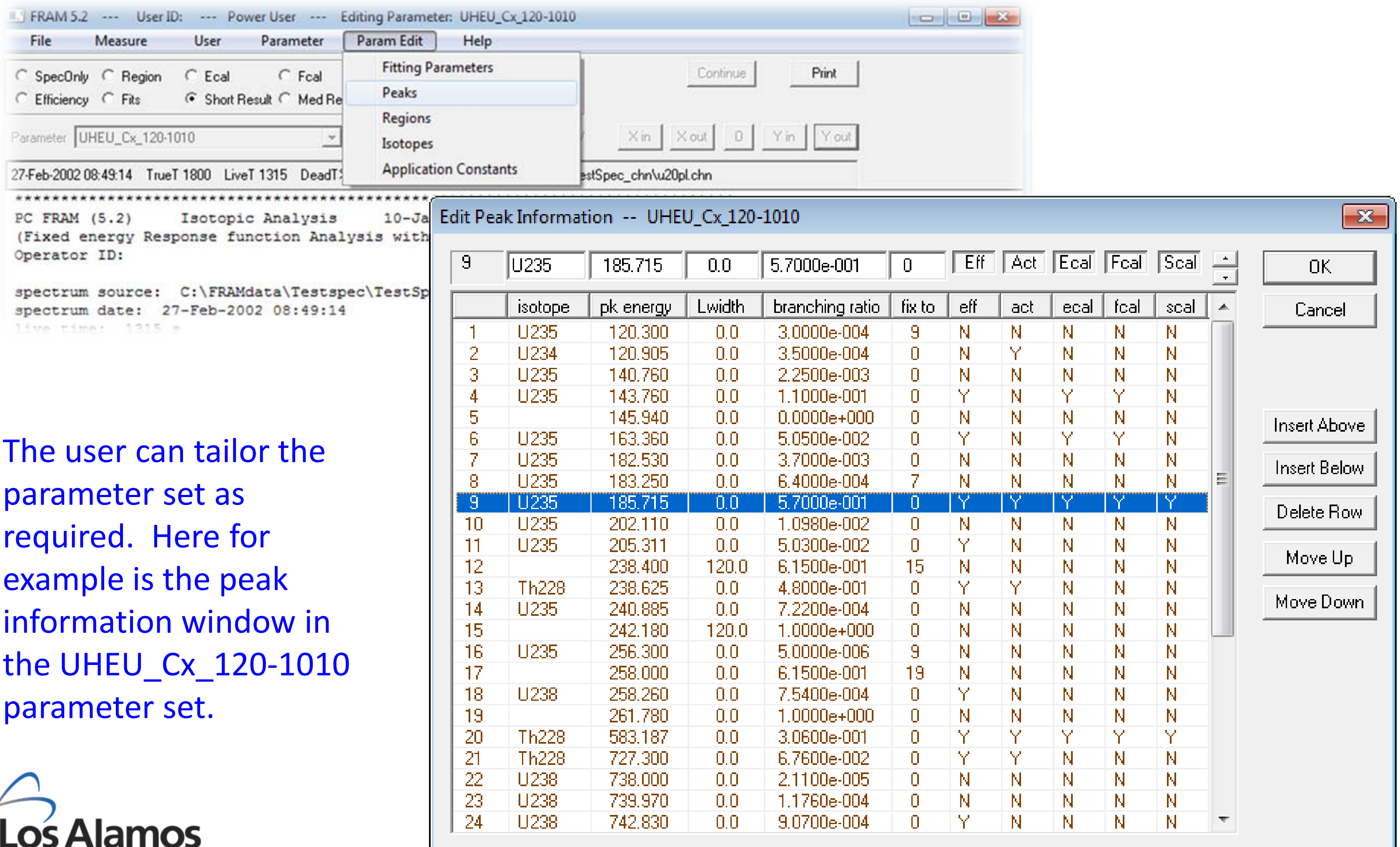
FRAM Analyze Window for Detective Data

Set Parameter set to one designed for a coaxial detector



Set default gain to 0.366 keV/ch

Editing Parameter Sets



The user can tailor the parameter set as required. Here for example is the peak information window in the UHEU_Cx_120-1010 parameter set.

Examples

- Uranium Examples:
  - Clean analysis of good spectrum
  - Analysis with incorrect gain
  - Enrichment Study (0.3, 20, 91%)
  - Shielded Item
- Plutonium Examples
  - Clean analysis of good spectrum
  - Analysis in different energy regions
  - Shielded Item
  - Correlation Function Limitations
  - Interferences / Additional Nuclides